1 590 870

PATENT SPECIFICATION (11) 1 590 870

(21) Application No. 24745/78 (22) Filed 31 May 1978
(61) Patent of Addition to No. 1 471 743 dated 29 July 1974
(44) Complete Specification published 10 June 1981
(51) INT CL³ C07D 261/14
(52) Index at acceptance
C2C 1173 1370 200 215 225 22Y 247 255 25Y 30Y 313 314 31Y 337 342 34Y 579 601 603 62X 805 80Y AA KF
(72) Inventors HISAJIRO YUKINAGA, SHINZABURO SUMIMOTO, ICHIRO ISHIZUKA and JITSUO SUGITA

(19)

ERRATUM

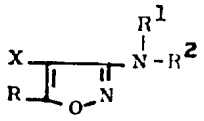
SPECIFICATION NO 1590870

Page 1, line 1 (71) *after* We, *insert* SHIONOGI SEIYAKU KABUSHIKI KAISHA known as

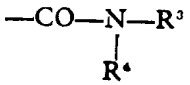
THE PATENT OFFICE
9 May 1983

Bas 251103/1

Our British Patent No. 1,471,743 describes and claims a compound represented by the formula:



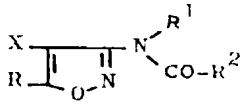
wherein R represents hydrogen, alkyl or aryl; R^1 represents hydrogen or alkyl; R^2 represents a group of the formula



wherein R^3 and R^4 each independently represent hydrogen, alkyl, alkenyl, alkynyl, aralkyl, aryl, alkoxy, or alkylthio, or R^3 and R^4 and the adjacent nitrogen atom form a ring optionally including another hetero atom (nitrogen, oxygen, or sulfur), or R^2 is a group of the formula —CO—Y—R^5 wherein R^5 represents alkyl, alkenyl, alkynyl, aralkyl, or aryl, and Y represents oxygen or sulfur; and X represents hydrogen, alkyl, or halogen; provided that R and X may optionally be taken together to represent alkylene, and alkyl, aralkyl, and/or aryl groups may each be optionally substituted with one or more substituents selected from halogen, alkyl, alkoxy, nitro or hydroxy. This invention can be regarded as a modification of the invention of said earlier patent in that the meanings for groups X and R are more restricted and in place of the group R^2 as defined in Patent No. 1,471,743 is to be found a group —CO—R^2 as defined below.

The isoxazole derivatives of the present invention show excellent herbicidal activity and exhibit very low toxicity towards humans, fish and other animals. It has also been discovered that the present isoxazole derivatives are smoothly decomposed or degraded in soil after their application thereto as herbicides.

According to the present invention there is provided a compound of the formula:



(I)

wherein R is t-butyl; R^1 is hydrogen; R^2 is alkyl (preferably C_2 to C_6), alkenyl (preferably C_3 or C_4) or cyclopropyl; and X is hydrogen or halogen.

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(72) Inventors HISAJIRO YUKINAGA, SHINZABURO SUMIMOTO, ICHIRO ISHIZUKA and JITSUO SUGITA

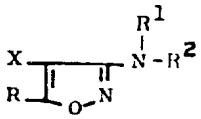
(19)

(54) N-(5-t-BUTYL-3-ISOXAZOLYL) ALKANAMIDE DERIVATIVES HAVING HERBICIDAL ACTIVITY

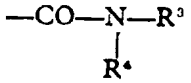
(71) We, SHIONOGI & CO. LTD., a Japanese Body Corporate, of 12 3-chome, Dosho-machi, Higashi-ku, Osaka, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to N-(5-t-butyl-3-isoxazolyl)alkanamide derivatives and to herbicidal formulations containing the same.

Our British Patent No. 1,471,743 describes and claims a compound represented by the formula:



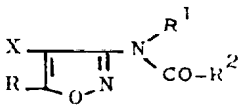
wherein R represents hydrogen, alkyl or aryl; R^1 represents hydrogen or alkyl; R^2 represents a group of the formula



wherein R^3 and R^4 each independently represent hydrogen, alkyl, alkenyl, alkynyl, aralkyl, aryl, alkoxy, or alkylthio, or R^3 and R^4 and the adjacent nitrogen atom form a ring optionally including another hetero atom (nitrogen, oxygen, or sulfur), or R^2 is a group of the formula —CO—Y—R^5 wherein R^5 represents alkyl, alkenyl, alkynyl, aralkyl, or aryl, and Y represents oxygen or sulfur; and X represents hydrogen, alkyl, or halogen; provided that R and X may optionally be taken together to represent alkylene, and alkyl, aralkyl, and/or aryl groups may each be optionally substituted with one or more substituents selected from halogen, alkyl, alkoxy, nitro or hydroxy. This invention can be regarded as a modification of the invention of said earlier patent in that the meanings for groups X and R are more restricted and in place of the group R^2 as defined in Patent No. 1,471,743 is to be found a group —CO—R^2 as defined below.

The isoxazole derivatives of the present invention show excellent herbicidal activity and exhibit very low toxicity towards humans, fish and other animals. It has also been discovered that the present isoxazole derivatives are smoothly decomposed or degraded in soil after their application thereto as herbicides.

According to the present invention there is provided a compound of the formula:



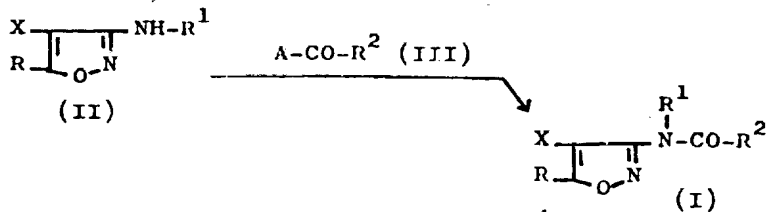
(I)

wherein R is t-butyl; R^1 is hydrogen; R^2 is alkyl (preferably C_2 to C_6), alkenyl (preferably C_3 or C_4) or cyclopropyl; and X is hydrogen or halogen.

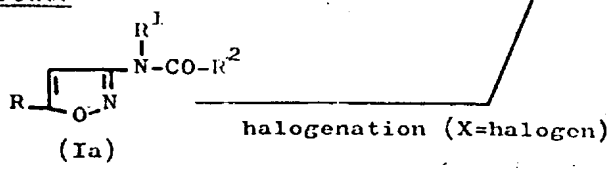
Among suitable alkyl groups R_2 are ethyl, propyl, i-propyl, butyl, i-butyl, t-butyl, pentyl and hexyl. Suitable alkenyl groups R^2 include allyl, isopropenyl, butenyl and butadienyl. Halogen may be, for example, chlorine or bromine.

The isoxazole derivatives (I) of the present invention can be prepared according to the following synthetic routes:

First route



Second route



wherein A is a residue of a reactive group such as halogen (e.g. chlorine or bromine) or an ester (e.g. tosyloxy, mesyloxy or —O—CO—R^2).

First route:

A compound (I) can be prepared by reacting an amine (II) with an acylating reagent (III) with or without the presence of a base (e.g. pyridine, triethylamine or sodium hydroxide) and in the presence or absence of an inert solvent (e.g. water, methanol, benzene, dimethylformamide or dimethylsulfoxide) at room temperature or with heating.

Second route:

Halogenation of an isoxazole (Ia) can be carried out in a conventional manner. Thus, an isoxazole (Ia) may be treated with a halogenating agent (e.g. chlorine, bromine or sulfuryl chloride) in an inert solvent (e.g. active acid, methylene chloride or chloroform) at room temperature or with heating.

The present invention includes a process for the preparation of a compound in accordance with the invention, which process is in accordance with the above routes.

Practical examples of the preparation of the present isoxazole derivatives (I) in accordance with each of the above routes are now given in the following Synthetic Examples.

Synthetic Example 1.

Propionic anhydride (5 ml) is added to 3-amino-5-t-butylisoxazole (2.52 g), and the resultant mixture is stirred at room temperature for 3 hours and then allowed to stand at room temperature overnight. The reaction mixture is poured onto icy water (50 ml). The precipitated crystals are filtered and shaken with benzene. The benzene layer is washed with saturated aqueous sodium bicarbonate and water each twice, dried over anhydrous sodium sulfate and evaporated to remove the solvent. The residue is recrystallized from cyclohexane to give N-(5-t-butyl-3-isoxazolyl)propionamide (2.91 g) is colourless crystals melting at 95.0° to 96.0° C.

Synthetic Examples 2 to 5.

The following products (Ia) are obtained from the corresponding amines (II) by reaction with the corresponding anhydride. $(R^2CO)_2O$, by procedures similar to that described in Example 1.

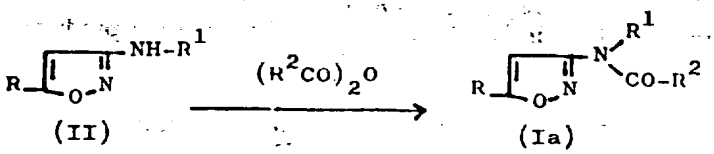


TABLE 1

Syn. Ex.	Product (Ia)			
	R	R^1	R^2	m.p. or b.p.
2	t-Bu	H	$-(CH_2)_2CH_3$	67.0–68.5°C
3	t-Bu	H	$-(CH_2)_3CH_3$	96.5–97.5°C
4	t-Bu	H	$-(CH_2)_4CH_3$	72.0–73.5°C
5	t-Bu	H	$-CH(CH_3)_2$	123.0–124.0°C

Note) The abbreviations in Table 1 have the following significance:

Bu (butyl), Pr (propyl), i-(iso-), t- (tertiary-), m.p. (melting point), b.p. (boiling point).

Synthetic Example 6.

To a solution of 3-amino-5-t-butylisoxazole (2.80 g) in pyridine (10 ml) is added dropwise isobutyryl chloride (2.34 g) keeping the mixture below 10° C. The reaction mixture is stirred with cooling for 30 minutes and at room temperature for 1 hour and evaporated to remove the pyridine. The residue is mixed with 5% hydrochloric acid solution (40 ml) and shaken with methylene chloride. The methylene chloride layer is separated, washed with saturated aqueous sodium bicarbonate and water, dried over anhydrous sodium sulfate and evaporated to remove the methylene chloride. The result is chromatographed on a column of silica gel and recrystallized from n-hexane to give N-(5-t-butyl-3-isoxazolyl)-isobutyramide (3.85 g) as colorless needles melting at 123.0 to 124.0° C.

Synthetic Examples 7 to 10.

Reactions are effected as in Example 6 to give the following products (Ia):

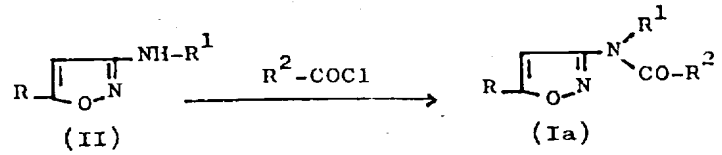
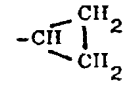


TABLE 2

Syn. Ex.	Product (Ia)			
	R	R^1	R^2	m.p. or b.p.
7	t-Bu	H	$-CH(CH_3)(CH_2CH_3)$	133.5–135.0°C
8	t-Bu	H	$-CH(CH_3)((CH_2)_2CH_3)$	96.5–97.5°C
9	t-Bu	H		146.0–147.0°C
10	t-Bu	H	$-C(CH_3)=CH_2$	70.5–71.5°C

Note) The abbreviations in Table 2 have the meanings given above for Table 1.

Synthetic Example 11.

Methylene chloride (20 ml) and sulfuryl chloride (5.40 g) are added to N-(5-t-butyl-3-isoxazolyl)cyclopropanecarboxamide (4.17 g) and refluxed with heating for 1.5 hours. The methylene chloride and the unreacted sulfuryl chloride are evaporated under reduced pressure. The residue is chromatographed on a column of silica gel and recrystallized from benzene to give N-(5-t-butyl-4-chloro-3-isoxazolyl)cyclopropanecarboxamide (4.10 g) as colorless needles melting at 129.5 to 131.0° C.

Synthetic Examples 12 to 17.

Reactions are effected as in Example 11 to give the following products (Ib):

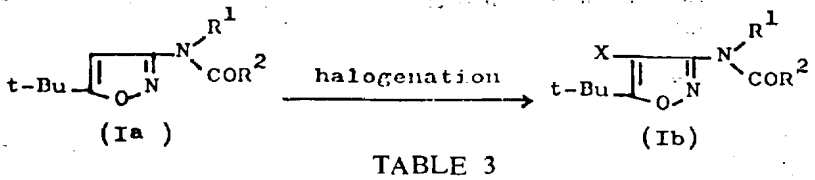
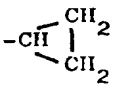


TABLE 3

Syn. Ex.	Product (Ib)			
	X	R^1	R^2	m.p.
12	Cl	H	$-CH_2CH_3$	88.0–89.0°C
13	Cl	H	$-CH(CH_3)_2$	96.5–97.5°C
14	Cl	H	$-CH(CH_3)CH_2CH_3$	99.0–100°C
15	Cl	H	$-(CH_2)_3CH_3$	80.0–81.5°C
16	Cl	H	$-CH(CH_3)(CH_2)_2CH_3$	92.0–93.0°C
17	Br	H		111.0–112.0°C

Note) * : Bromine and 1,2-dichloroethane are used.

Experiment 1.

a) Compounds tested:

Compound No.	Compound
1	N-(5-t-butyl-3-isoxazolyl)propionamide
2	N-(5-t-butyl-3-isoxazolyl)isobutyramide
3	N-(5-t-butyl-3-isoxazolyl)-sec-valeramide
4	N-(5-t-butyl-3-isoxazolyl)valeramide
5	N-(5-t-butyl-3-isoxazolyl)-2-methylvaleramide
6	N-(5-t-butyl-3-isoxazolyl)hexanamide
7	N-(5-t-butyl-3-isoxazolyl)-2-methylhexanamide
8	N-(5-t-butyl-3-isoxazolyl)cyclopropanecarboxamide
9	PCP—Na (Sodium pentachlorophenoxide)

b) Test method:

(1) Pre-emergence test:

25 seeds of a test plant were sown in sandy soil in a polyethylene cup (diameter: 9 cm). After sowing, the seeds were covered with sandy soil to about 5 mm depth and an aqueous suspension of a test compound at a concentration of 100 ppm using Tween 20 (trademark of Atlas Powder Co.) as a spreader was applied over the surface of the sandy soil. The application rate of the test compound was 10 g/are and 30 g/are in aqueous suspension (water dilution: 10 L/are) applied by a sprayer. Administration was effected at 25° C in a greenhouse in natural sunlight. The degree of germination was evaluated 3 weeks after the application.

(2) Post-emergence test:

A test compound was applied to young plants 10 days after seeding. Administration and evaluation were effected as described above (1).

c) Method of evaluation:

The number of surviving plants was determined by the naked eye and the survival percentage for the sown seeds was then calculated. The survival percentage was marked in six degrees as follows:

Survival percentage of the plant tested	Mark
Not more than 10%	5
11—25%	4
26—50%	3
51—75%	2
76—90%	1
Not less than 91%	0

d) Result:

TABLE 4.

Compound No.	Application rate (g/are)	Herbicidal Activity											
		Pre-emergence test						Post-emergence test					
		A	B	C	D	E	F	A	B	C	D	E	F
1	10	0	0	0	4	4	1	0	0	0	5	4	5
	30	0	2	3	5	5	5	0	1	2	5	5	5
2	10	0	3	5	5	5	5	0	1	5	5	5	5
	30	0	4	5	5	5	5	0	5	5	5	5	5
3	10	0	0	1	0	0	0	0	0	0	4	0	0
	30	0	3	4	4	4	1	0	0	5	5	5	3
4	10	0	0	2	2	2	2	0	1	4	3	4	5
	30	0	4	5	5	5	5	0	2	5	5	5	5
5	10	0	3	5	5	5	5	0	0	5	2	5	5
	30	0	5	5	5	5	5	0	3	5	5	5	5
6	10	0	0	2	3	1	1	0	1	5	5	3	3
	30	0	0	2	5	2	0	0	1	5	5	5	3
7	10	0	0	5	5	5	5	0	0	2	5	5	5
	30	0	4	5	5	5	5	0	0	2	5	5	5
8	10	0	5	5	5	5	5	0	5	5	5	5	5
	30	0	5	5	5	5	5	3	5	5	5	5	5
9	10	0	0	0	0	2	3	0	0	0	0	2	5
	30	0	0	0	0	2	3	0	0	0	0	4	5
	50	0	2	2	0	5	5	0	0	4	2	5	5

Note) The abbreviations each have the following significance:

A (*Triticum aertivum*, B (*Echinochloa crusgalli*),
C (*Digitaria adscendens*), D (*Brassica campestris*),
E (*Polygonum logisetum*), F (*Amaranthus retroflexus*),

Thus, it can be seen that the tested amides (I) (Compounds Nos. 1 to 8) each exhibited hardly any herbicidal activity against *Triticum aestivum* but showed excellent and selective herbicidal activity against *Echinochloa crusgalli, Digitaria adscendens, Brassica campestris, Polygonum longisetum* and *Amaranthus retroflexus*, and their herbicidal activities were far more potent than that of a commercially available herbicide, PCP—Na (Compound No. 9).

Experiment 2.

The toxicity of N-(5-t-butyl-3-isoxazolyl)isobutyramide was examined on *Oryzias latipes* and found to be very low in comparison with that of commercially available PCP—Na.

Experiment 3.

Herbicidal activity against *Echinochloa crusgalli* and *Monochoria vaginalis* in water-pooled paddy fields.

a) Compound tested:

Compound No.	
1	N-(5-t-butyl-3-isoxazolyl)-2-methylpentanamide
2	N-(5-t-butyl-3-isoxazolyl)cyclopropanecarboxamide
3	Propanil (Control)

b) Test method:

Into 2×10^{-4} are of a Wagner pot was placed paddy field soil and water was poured in to a depth of 3 cm. 25 seeds of a test plant were sown. Test compound was applied to *Echinochloa crusgalli* at the time of pre-emergence, one leaf term, two leaves term and three leaves term and to *Monochoria vaginalis* at the time of pre-emergence, small leaf term, one leaf term and two leaves term. The application rate of the test compounds included 6.25 g, 12.5 g, 25 g, 50 g, 75 g and 100 g each per are. The number of surviving plants was examined in comparison with that in a non-applied section. The evaluation was marked in six degrees of survival percentage as in Experiment 1.

d) Result:

TABLE 5

Compound No.	Application rate (g/are)	*Echinochloa crusgalli*				*Monochoria vaginalis*			
		PRE	1L	2L	3L	PRE	SL	1L	2L
1	6.25	2	1	1	0	0	4	3	3
	12.5	2	3	3	0	0	5	4	3
	25	2	5	5	0	0	5	5	4
	50	3	5	5	2	2	5	5	5
	75	5	5	5	2	3	5	5	5
	100	5	5	5	4	3	5	5	5
2	6.25	3	5	5	5	4	5	5	5
	12.5	5	5	5	5	5	5	5	5
	25	5	5	5	5	5	5	5	5
	50	5	5	5	5	5	5	5	5
	75	5	5	5	5	5	5	5	5
	100	5	5	5	5	5	5	5	5
3	6.25	0	0	0	0	0	2	1	1
	12.5	1	1	0	0	0	2	2	1
	25	3	2	1	0	0	2	2	2
	50	4	2	1	0	0	3	3	3
	75	4	3	2	0	2	4	3	3
	100	5	3	2	0	2	4	3	3

Note) The abbreviations each have the following significance:

PRE (pre-emergence), IL (one leaf term), 2L (two leaves term), 3L (three leaves term), SL (small leaf term).

d) Conclusion:

Thus, the tested compounds of this invention (Compounds Nos. 1 and 2) showed excellent herbicidal activity against *Echinochloa crusgalli* and *Monochoria vaginalis* in water-pooled paddy fields in comparison with a commercially available herbicide, propanil. Additionally, chemical poisoning of rice plants by any of the compounds (Compound Nos. 1—3) was almost never observed.

The present isoxazole derivatives (I) show excellent herbicidal activity against various grasses with a small rate of application. These compounds can also be used as non-selective or selective herbicides by changing the rate of application thereof. The herbicides of this invention are generally applicable to various crops including wheat, barley, corn, carrots, peanuts, peas and rice plants in order to protect them from undesired weeds and grasses. They can also be applied to sugar cane, potatoes, sweet potatoes, mentha, egg-plant or Spanish paprica after planting thereof. Virtually no harmful chemical effect at all is observed as a result of the action of the present herbicides on these crops, such effects as are observed being so slight that the crops easily recover. Furthermore, the present herbicides are quite harmless to humans and domestic

animals, and they also exhibit very low toxicity towards fish and shell fish. Thus, the herbicides of the present invention have a very high degree of safety and also result in a suitable amount of residual active compound remaining in the soil after use thereof.

Thus, the present invention includes a herbicidal formulation which comprises a compound in accordance with the invention formulated for herbicidal use and, optionally, together with a diluent, carrier or excipient.

The present herbicidal formulations may, for example, be prepared by mixing the effective isoxazole derivative (I) with an inert solid or liquid carrier, optionally in combination with a further adjuvant (e.g. selected from emulsifiers, stabilizers, dispersants, suspending agents, spreaders, penetrants and wetting agents) and converting the resultant mixture into the desired preparation form (e.g. an emulsion, a wettable powder, granules, a dust, tablets or pills). Examples of suitable carriers are solid carriers (e.g. clay, talc, diatomaceous earth or bentonite) and liquid carriers (e.g. water, alcohols, acetone, benzene, toluene, xylene, solvent naphtha or cyclohexane).

The herbicidal formulations of this invention may also include, and the present compounds may be used with, agricultural chemicals (e.g. insecticides, fungicides or other herbicides), manuring ingredients (e.g. ammonium sulfate or urea) or soil treating agents.

The invention thus includes a method of producing a herbicidal effect in an environment, which method comprises administering to the environment an effective amount of a compound or formulation of the invention. Such a method may be used in treating crop areas to remove or inhibit the growth of weeds or grasses.

The invention also includes a method of killing a plant, which method comprises applying thereto an effective amount of a compound or formulation in accordance with the invention.

The herbicidal formulations of the present invention will now be illustrated by the following Examples.

Example A.

N-(5-t-butyl-3-isoxazolyl)isobutylamide (50 parts by weight), calcium ligninsulfonate (3 parts by weight), Sorpol (made by Toho Chemical Industry, Co., Ltd.) (3 parts by weight) and diatomaceous earth (44 parts by weight) are mixed and pulverised to produce a wettable powder.

Example B.

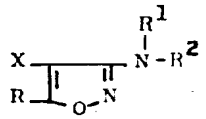
N-(5-t-butyl-3-isoxazolyl)-2-methylvaleramide (5 parts by weight) and clay (95 parts by weight) are mixed and pulverized to produce a dust.

Example C.

N-(5-t-butyl-3-isoxazolyl)cycloprapanecarboxamide (5 parts by weight), calcium ligninsulfonate (5 parts by weight), bentonite (30 parts by weight) and clay (60 parts by weight) are mixed, pulverized, mixed with water, kneaded, granulated and dried to produce granules.

WHAT WE CLAIM IS:—

1. A compound of the formula:



(I)

wherein R is t-butyl; R^1 is hydrogen; R^2 is alkyl, alkenyl or cyclopropyl; and X is hydrogen or halogen.

2. A compound as claimed in claim 1, wherein R^2 is (C_2 to C_6)alkyl or (C_3 or C_4)alkenyl.

3. A compound as claimed in claim 2, wherein R^2 is ethyl, propyl, i-propyl, butyl, t-butyl, pentyl, hexyl, allyl, isopropyl, butenyl or butadienyl.

4. A compound as claimed in any one of claims 1 to 3 wherein X is chlorine or bromine.

5. N-(5-t-butyl-3-isoxazolyl)propionamide.

6. N-(5-t-butyl-3-isoxazolyl)isobutyramide.

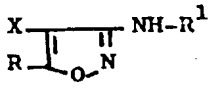
7. N-(5-t-butyl-3-isoxazolyl)-sec-valeramide.

8. N-(5-t-butyl-3-isoxazolyl)valeramide.

9. N-(5-t-butyl-3-isoxazolyl)-2-methylvaleramide.
10. N-(5-t-butyl-3-isoxazolyl)hexanamide.
11. N-(5-t-butyl-3-isoxazolyl)-2-methylhexanamide.
12. N-(5-t-butyl-3-isoxazolyl)cyclopropanecarboxamide.

13. A compound as claimed in claim 1 and substantially as referred to hereinbefore, other than a compound as claimed in any one of claims 5 to 12.

14. A process for preparing a compound as claimed in claim 1, which process comprises either a) reacting an amine of the formula:

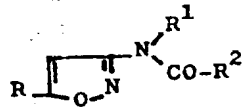


(II)

with an acylating reagent of the formula:

$$A—CO—R^2 \qquad (III)$$

wherein A is a residue of reactive group; or b) halogenating an isoxazole of the formula:



(Ia)

15. A process as claimed in claim 14, wherein A is halogen or an ester group.

16. A process as claimed in claim 14 or claim 15, wherein, in (a), the reaction is effected in the presence of a base.

17. A process as claimed in claim 16, wherein the base is pyridine, triethylamine or sodium hydroxide.

18. A process as claimed in any one of claims 14 to 17, wherein in (a), the reaction is effected in an inert solvent.

19. A process as claimed in claim 14, wherein the halogenation is achieved by the use of chlorine, bromine or sulfuryl chloride as a halogenating agent.

20. A process as claimed in claim 14 or claim 19, wherein the halogenation is effected in an inert solvent.

21. A process as claimed in claim 14 and substantially as hereinbefore described in any one of the foregoing Synthetic Examples.

22. A compound as claimed in claim 1 which has been prepared by a process as claimed in any one of claims 14 to 21.

23. A herbicidal formulation which comprises a compound as claimed in any one of claims 1 to 13 and 22 formulated for herbicidal use.

24. A formulation as claimed in claim 23 also comprising a diluent, carrier or excipient.

25. A formulation as claimed in claim 24, wherein the diluent, carrier or excipient is clay, talc, diatomaceous earth, bentonite, water, an alcohol, acetone, benzene, toluene, xylene, solvent, naphtha or cyclohexane.

26. A formulation as claimed in any one of claims 23 to 25 also comprising an emulsifier, at stabiliser, a dispersant, a suspending agent, a spreader, a penetrant, a wetting agent, an insecticide, a fungicide, a herbicide other than a compound as claimed in any one of claims 1 to 13 and 22, a manuring ingredient or a soil treating agent.

27. A formulation as claimed in any one of claims 23 to 26 in the form of an emulsion, a wettable powder, granules, a dust, a pill or a tablet.

28. A formulation as claimed in claim 23 and substantially as hereinbefore described in any one of Examples A to C.

29. A method of producing a herbicidal effect in an environment, which method comprises administering to the environment an effective amount of a compound as claimed in any one of claims 1 to 13 and 22 or a formulation as claimed in any one of claims 23 to 28.

30. A method as claimed in claim 29 when used in treating a crop area to remove or inhibit the growth of weeds or grasses.

31. A method as claimed in claim 30, wherein the crop is wheat, barley, corn, carrots, peanuts, peas, rice, sugar cane, potatoes, sweet potatoes, mentha, egg-plant or Spanish paprica.

32. A method as claimed in claim 29 and substantially as hereinbefore described in Experiment 1 or Experiment 3.

33. A crop which has been grown in an area to which a method as claimed in claim 30 or claim 31 has been applied.

34. A method of killing a plant, which method comprises applying thereto an effective amount of a compound as claimed in any one of claims 1 to 13 and 22 or of a formulation as claimed in any one of claims 23 to 28.

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